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13. ABSTRACT (Maximum 200 words) The main objective of our research under this DURIP program was the evaluation of new or improved optical materials currently under development either at CREOL, or in industry through collaboration with CREOL. DURIP support was used to purchase laser diodes and related equipment for pumping a variety of solid state materials which show promise for laser and phosphor applications. These include the laser candidates Tm,Dy:BYF, Tm,Ho:BYF, Yb,Pr:NYF, and Nd:NYF as well as several NYF-based phosphors sensitized by Yb. Experiments using laser diodes as the pump source are essential for a complete characterization of these materials, which exploit the advantages of diode-pumping, leading potentially to compact, efficient, and rugged systems. Much of the supported research was accomplished in partnership with DoD laboratories and contractors who supported the growth of the subject materials. An additional accomplishment of this DURIP program was its significant contribution to the education of the graduate and undergraduate students involved in the research. This research is described in the Ph.D. theses of two students, in addition to several publications in the literature.					
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Office of Research

June 23, 2000

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Administrative Contracting Officer
Department of the Air Force
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Subject Contract No. F49620-97-1-0433

Dear Ms. Veon:

On behalf of Dr. Hans Jenssen, we have enclosed the Final Report for the subject Contract for your review and files. We apologize for the delay in this matter and any inconvenience it may have caused.

If you have any questions or need additional information, please contact me at (407) 823-3062 or email at kmyers@mail.ucf.edu.

Sincerely,

A handwritten signature in ink, appearing to read 'Kim Myers'.

Kim Myers
Sr. Contract Specialist

Enclosure

cc: Dr. H. Jenssen
Mr. M. Wagenhauser

klm

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Introduction

The support provided by this DURIP program was used to purchase laser diodes, power supplies for these diode lasers, optics and optical mounts. This enabled our group to expand its capabilities in evaluating new or improved laser materials that were developed at CREOL, or in the industry, in collaboration with CREOL. Another benefit was in education by involving graduate students in this research.

The requirements on a laser material are quite different when laser diodes are used for pumping rather than conventional lamp pumping. The two most striking differences are that diode pumping can be done at a wavelength matched to the absorption of the laser material, with relatively small difference between pump energy and laser photon energy, and that the intensity of the pump light can be much higher. Ever increasing power density available from the diode laser arrays used for pumping has led to as severe load (in power/cm³) on the laser material as achieved with lamp pumping although the distribution of the load may be quite different.

The laser diodes purchased in this program were utilized to pump a variety of available solid state laser materials, that showed promise of being able to be used in compact, rugged laser systems.

Objectives

Our objective in the research performed with the equipment purchased under this DURIP program was to work in partnership with DoD laboratories and contractors to develop and improve solid state laser materials designed for laser diode pumping. We would then be able to

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provide more completely characterized material, in a more timely manner, with the added value of realistic diode laser pumped laser test data.

We have had several collaborations/programs with DoD laboratories directly or through sub-contracts via small business. A variety of laser crystals were evaluated, as well as glass and phosphor materials.

New fluoride materials such as BaY_2F_8 and NaYF_4 , due to their low phonon energies have great potential for laser diode pumped lasers. Dr. Jenssen and his graduates students, Rita Peterson and Anna Tabirian have evaluated several rare-earth-doped fluoride crystals grown at AC Materials, Winter Park, FL. Support for the students and P.I. was provided by Schafer Corporation, by AC Materials and Lasergenics, through SBIR sub-contracts to CREOL.

A 968 nm diode laser purchased in this program was used successfully to pump a variety of $\text{Er}:\text{BYF}$ lasers. This work was done in collaboration with Schafer Corporation that supported a graduate student. Crystal samples representing a variety of crystallographic orientations and Er concentrations were lased in the same resonator in order to determine the optimal material parameters for lasing. An important lesson learned during this project was that power output of the diode laser is only one part of the requirements for successful diode pumping. The beam quality, or the brightness of the output is equally or more important.

A 792 nm diode laser purchased in this program was used successfully by Dr. Jenssen and a graduate student, Rita Peterson to demonstrate lasing of a new crystal, $\text{Nd}:\text{NaYF}_4$. The study was funded by AC Materials under a NSF SBIR Phase I subcontract to CREOL. As part of the same subcontract, a new material, $\text{Pr},\text{Yb}:\text{NaYF}_4$ was evaluated as a candidate for amplifier for

telecommunications. The spectroscopic evaluation indicates that this material is an excellent candidate for diode-pumped laser emitting at 1300 nm. This wavelength is important for telecommunications since nearly all existing transmission systems currently use the 1300 nm band in which EDFAs do not operate. Current Pr^{3+} doped amplifiers are nowhere as good as EDFAs with gains of only 12dB and restricted bandwidths. This new material has a bandwidth of over 100 nm, and this will greatly enhance the utility of Pr^{3+} as a rare-earth ion for telecommunications. The laser evaluation will be continued as part of a NSF Phase II SBIR subcontract to CREOL.

Phosphor materials based on rare-earth-doped NaYF_4 powder and prepared at AC Materials were evaluated by Prof. Bass and his Post-doctoral student, Alexandra Rapaport. Visible emission spectra of various dopants were recorded after pumping ytterbium with a cw diode laser at 968 nm. The infrared light was also scanned and modulated in order to obtain a demonstration of 1D display, under an ARO program. The phosphor powders were excited by diodes purchased under this DURIP program. The results were positive, so that continuation of the program under a NSF SBIR Phase II will start in the near future.

A 792 nm diode laser purchased in this program was used by Dr. Jenssen and Rita Peterson to attempt obtaining lasing in a new crystal, $\text{Tm,Dy:BaY}_2\text{F}_8$. This study was funded by AC Materials under a BMDO Phase I subcontract to CREOL. The crystal studied is a candidate for 3 μm laser and the lasing at room temperature has not been achieved yet. Modeling suggests that the power densities available using the 792 nm diode

laser should be more than sufficient, but in all likelihood the crystal did not contain a high enough active ion concentration to reach laser threshold.

Dr. Jenssen and his graduate students, under support from Lasergenics, through a BMDO SBIR Phase I subcontract, initiated evaluation of a Ho:BYF crystal as a candidate for a 4 μm laser. Recently, Dr. Jenssen, and his graduate students, Scott Buchter (now at MIT Lincoln Laboratory) and Anna Tabirian demonstrated lasing at 3.98 μm using a Cr:LiSAF laser for pumping. Although the plan was to use a diode laser as a pump for this laser scheme this has not yet been attempted.

One graduate student, Robert Hopkins was involved in the evaluation of a 2 μm laser based on Tm,Ho: BaY₂F₈, and Tm,Ho: NaYF₄ and worked at the Wright Laboratory at Eglin AFB under an AFOSR sponsored summer graduate student program. As a result of this summer program, Air Force Captain Ken Dinndorf later spent 2 weeks at CREOL and with students used the laser diodes for laser testing of these crystals. The results were reported at OSA TOPS Vol. 26, Advanced Solid State Lasers, 1999.

Prof. Bass and his graduate students investigated neodymium doped and ytterbium doped orthophosphate crystals. These crystals were grown at Oak Ridge. Specific spectroscopic properties like the stimulated emission cross-sections were obtained by excitation of the active ion by a cw diode laser. Lasing was then tested and obtained by end-pumping the most promising crystals with the 1 W single emitter laser diode at 804 nm.

A new optical amplifier for the 1.3-1.6 μm region was investigated by Prof. Bass and his graduate student, Corey LaPine, undergraduate student, Stephanie Barre along with Post Doctoral Fellow, Alexandra Rapaport. The

diodes at 970 and 804 nm were used as pumps to excite the active ions so that they might amplify light in the 1.3 to 1.5 μm range. Earlier research on the fluorescence spectrum of tetravalent chromium doped aluminate glass suggested that it could serve as the desired amplifier. The glass was prepared at CREOL.

Another project in progress, by Prof. Bass and students is the study of the temperature dependence of the stimulated emission cross section of Nd:YAG. Recent modeling of the temperature dependence of the performance of passively and actively Q-switched lasers along with measurements of the temperature dependence of the optical properties of several laser cavity components have shown that the critical temperature dependent quantity is the temperature coefficient of the stimulated emission cross section. Prof. Bass and students have initiated measurements of this quantity for Nd:YAG since literature searches have not revealed any trustworthy values. The diode laser at 804 nm is used in this work to excite the Nd fluorescence while the spectral shape of the emission is measured as a function of temperature. The temperature range of interest is that of practical laser systems, e.g. from ~ -60 to $\sim +80^\circ\text{C}$. An undergraduate student, Andrew Howard, and a graduate student, Eric Nelson work on this project.

Research employing the equipment purchased under this DURIP program is described in several papers, reports to SBIR sponsors and in the Ph.D. thesis of Guohua Xiao and Rita Peterson.

"Two Micron Diode-Pumped Laser Operation of $\text{Tm,Ho:BaY}_2\text{F}_8$," K. Dinndorf and H. Miller, A. Tabirian and H. P. Jenssen, and A. Cassanho, in OSA TOPS, Vol26, Advanced Solid State Lasers, Martin Fejer et al, eds., page 506,1999

"Optical spectroscopy and lasing properties of neodymium-doped lutetium orthophosphate", A. Rapaport, O. Mouteau, M. Bass, L. A. Boatner, and C. Deka, in JOSA B 16 , 911 (1999)

"Optical spectroscopy of erbium-doped lutetium orthophosphate", ", A. Rapaport, V. David, M. Bass, C. Deka, and L. A. Boatner, in Journal of Luminescence 85 , 155 (1999)

Attached are the first pages of the published papers and front page of Xiao's thesis.

Optical spectroscopy and lasing properties of neodymium-doped lutetium orthophosphate

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Studies of the spectroscopic and lasing properties of Nd³⁺-doped LuPO₄ are reported. LuPO₄ permits incorporation of lanthanide dopants in high concentration. We report the absorption and luminescence properties of Nd³⁺ ions doped at 10% in this host. Promising laser performance with diode laser pumping is also described. © 1999 Optical Society of America (S0740-3224(99)00806-1)

OCIS codes: 140.5680, 160.5690, 300.6280, 140.3480, 140.3380, 160.3380.

1. INTRODUCTION

Diode-pumped microchip lasers are of interest because of their potential to be small, sturdy, and high-efficiency devices.¹ A key element in an improved Nd³⁺-doped microchip laser would be a host material that would permit incorporation of laser-active ions at a higher concentration than is possible in yttrium aluminum garnet (~1%). Host crystals having the general chemical formula RPO₄, where R is a lanthanide element, have been shown to be able to accommodate various lanthanide ions as dopants in large concentration without compromising the crystal-line structure. As a result, we have initiated studies of the spectroscopic and lasing properties of these materials. In this paper we report on Nd³⁺-doped LuPO₄ (Nd:LuPO₄).

Lanthanide orthophosphates are characterized by an unusual combination of favorable chemical and physical properties. As a result, a great deal of attention has been given to these materials as potential nuclear-waste-containment hosts. Interest has recently been shown in their optical, magnetic, structural, and electronic properties since the orthophosphates have proven to be very stable and radiation-damage-resistant materials. Therefore these materials may have potential applications as thermophosphors for remote temperature measurements, as x-ray and γ-ray scintillators for medical imaging applications, and, as in the present work, as laser hosts.

2. MATERIAL PROPERTIES

Lutetium orthophosphate is a uniaxial crystal with the optical axis along the crystallographic *c* axis. The crystal structure is a simple body-centered tetragonal (space

group *I41/amd*). Lanthanide ions doped into the subject host substitute for the Lu³⁺ ions, occupying sites of unique point-group symmetry *D*_{2d}. Lanthanide orthophosphates are extremely durable materials that are not attacked by most chemicals, including boiling nitric and hydrochloric acid. Its melting temperature is 2000 °C. The refractive index of the material in this paper is taken to be 1.7.²⁻⁴ Reference 2 gives the ordinary and extraordinary indices in LuPO₄ as 1.694 and 1.728, respectively. However, Ref. 4 gives a value of 1.62 for the index of refraction of the stoichiometric material NdPO₄, suggesting that the index of a 10% Nd³⁺:LuPO₄ crystal lies between 1.62 and 1.73. LuPO₄ has a Moh hardness coefficient between 4 and 7.5.

3. CRYSTAL GROWTH

We grew lutetium orthophosphate crystals by dissolving and reacting lutetium oxide in molten-lead pyrophosphates (PbHPO₄) in a platinum crucible. A mixture of 3.5 g of lutetium oxide, Lu₂O₃, and 60 g of PbHPO₄ was placed in a 50-cm³ platinum crucible covered with a tight-fitting Pt lid in order to reduce flux evaporation. Decomposition of PbHPO₄ at high temperature resulted in the formation of Pb₂P₂O₇ that served as the flux. The oxide dopant Nd₂O₃ was then added to this melt. The Pt crucible was placed in a furnace, heated at 1360 °C for 16 h, and then slowly cooled to 900 °C at a rate of one degree per hour. At that point the furnace was allowed to cool to room temperature, and we removed the orthophosphate crystals by dissolving the flux in boiling nitric acid. In the present spectroscopic and lasing measurements we

THE DESIGN OF PASSIVELY Q-SWITCHED SOLID STATE LASERS

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A DISSERTATION

Submitted in partial fulfillment of the requirements
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Two Micron Diode-Pumped Laser Operation of Tm, Ho:BaY₂F₈

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Abstract: Diode-pumped Tm, Ho:BaY₂F₈ was operated as a room-temperature two micron laser. Laser results and various spectroscopic parameters are reported.

OCIS codes: (140.3380) Laser materials; (140.5680) Rare earth and transition metal solid-state lasers

Introduction

BaY₂F₈ is a promising host material for applications in solid-state laser systems. It is a birefringent material that is optically transparent from its infrared absorption edge at 9 μm to beyond 200 nm in the ultraviolet[1]. Its phonon spectra is relatively low energy, with a maximum of 415 cm^{-1} [2], and it has low non-radiative relaxation rates relative to garnets[3]. These characteristics make it potentially attractive for mid-infrared energy storage applications.

Crystal Structure

BaY₂F₈ crystallizes in the β -BaTm₂F₈ structure and is isostructural with BaLn₂F₈, with Ln \rightarrow {Er, Tm, and Yb}. This structure is a monoclinic basis with unit cell parameters $a=6.935\pm0.001\text{\AA}$, $b=10.457\pm0.002\text{\AA}$, $c=4.243\pm0.001\text{\AA}$, and $\beta=99^\circ40'\pm2'$. [4] The unit cell has symmetry $C_{2/m}$ and contains 2 formula units of BaY₂F₈. Rare earths substitutionally enter the Y³⁺ site (symmetry C₂). BaDy₂F₈ and BaHo₂F₈ are also isostructural with BaY₂F₈, but some problems have been experienced with high concentration dopings of these rare-earths in BaY₂F₈[5]. Other rare earths can be substituted into the system in small concentrations.

Optical properties of BaY₂F₈

BaY₂F₈ is a biaxial crystal; due to its monoclinic structure, the optical axes (x,z) in the a-c plane need not coincide with the crystallographic axes. Symmetry constrains the y optical axis to coincide with the b crystallographic axis[6]. Three different indices of refraction are needed to describe the dielectric properties of BaY₂F₈ as well as an angle, ρ , describing the angular difference between the orientation of the optical axes and the crystallographic axes of the crystal. At 2 μm , these values are $n_x=1.51$, $n_y=1.52$, $n_z=1.50$, and $\rho=20^\circ$ [1]. Figure 1 displays the relationship between the crystallographic and optical axes of BaY₂F₈.

Similar symmetry constraints apply to the imaginary portion of the dielectric tensor. The angular orientation of the principle axes of the imaginary portion of the dielectric tensor (which corresponds to the minima and maxima of the transition cross-sections) has been experimentally determined to differ from the crystallographic and optical axes in the a-c plane of this crystal structure[7].

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Optical spectroscopy of erbium-doped lutetium orthophosphate

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